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Satoshi SEO et al.

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1793

Title of the Invention:

ORGANIC-INORGANIC HYBRID MATERIAL,

COMPOSITION FOR SYNTHESIZING THE SAME, AND

MANUFACTURING METHOD OF THE SAME

#### VERIFICATION OF TRANSLATION

Commissioner for Patents P.O.Box 1450 Alexandria, VA 22313-1450

Sir:

I, Ryoji Nomura, C/O Semiconductor Energy Laboratory Co., Ltd. 398, Hase, Atsugi-shi, Kanagawa-ken 243-0036 Japan, a translator, herewith declare:

that I am well acquainted with both the Japanese and English Languages;

that I am the translator of the attached English translation of the Japanese Patent Application No. 2003-085688 filed on March 26, 2003; and

that to the best of my knowledge and belief the following is a true and correct English translation of the Japanese Patent Application No. 2003-085688 filed on March 26, 2003.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: April 29, 2008

Name: Ryoji Nomura

[Name of Document] Patent Application [Reference Number] P007059 [Filing Date] March 26, 2003 [Attention] Commissioner, Patent Office [Inventor] 5 [Address] 398, Hase, Atsugi-shi, Kanagawa-ken c/o Semiconductor Energy Laboratory [Name] Satoshi SEO [Inventor] 10 [Address] 398, Hase, Atsugi-shi, Kanagawa-ken c/o Semiconductor Energy Laboratory [Name] Harue NAKASHIMA [Inventor] [Address] 398, Hase, Atsugi-shi, Kanagawa-ken 15 c/o Semiconductor Energy Laboratory [Name] Ryoji NOMURA [Applicant] [Identification Number] 000153878 [Name] Semiconductor Energy Laboratory 20 [Representative] Shunpei YAMAZAKI [Indication of Handlings] [Number of Prepayment Note] 002543 [Payment Amount] ¥21,000 [List of Attachment] 25 [Attachment] Scope of Claims 1 Specification [Attachment] 1 [Attachment] Drawing 1 [Attachment] Abstract 1

Requested

[Proof]

[Document Name]

Specification

[Title of Invention]

ORGANIC-INORGANIC

HYBRID

MATERIAL,

COMPOSITION FOR SYNTHESIZING THE SAME, AND

MANUFACTURING METHOD OF THE SAME

5 [Scope of Claims]

[Claim 1]

An organic-inorganic hybrid material having a metal oxide matrix having one kind or plural kinds of metal atoms and a ligand bonded to the metal atom by chelating, wherein the ligand shows a coloring property by chelating the metal atom.

10 [Claim 2]

An organic-inorganic hybrid material having a metal oxide matrix having one kind or plural kinds of metal atoms and a ligand bonded to the metal atom by chelating, wherein the ligand shows a light-emitting property by chelating the metal atom.

[Claim 3]

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An organic-inorganic hybrid material having a metal oxide matrix having one kind or plural kinds of metal atoms and a ligand bonded to the metal atom by chelating, wherein the ligand shows semiconductivity by chelating the metal atom.

[Claim 4]

An organic-inorganic hybrid material having a metal oxide matrix having one kind or plural kinds of metal atoms and a ligand in which an oxygen atom from a phenolic hydroxyl group is bonded to the metal atom, wherein the ligand further has a heterocycle with a nitrogen atom as a hetero atom and chelates the metal atom by the oxygen atom and the nitrogen atom.

[Claim 5]

An organic-inorganic hybrid material having a metal oxide matrix having one kind or plural kinds of metal atoms and a ligand in which an oxygen atom from a phenolic hydroxyl group is bonded to the metal atom, wherein the ligand further has a carbonyl group and chelates the metal atom by the oxygen atom and the carbonyl group.

[Claim 6]

An organic-inorganic hybrid material having a metal oxide matrix having one kind or plural kinds of metal atoms, and a ligand in which an oxygen atom from a phenolic hydroxyl group is bonded to the metal atom, wherein the ligand further has an azomethine group and chelates the metal atom by the oxygen atom and the azomethine group.

### [Claim 7]

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An organic-inorganic hybrid material having a metal oxide matrix having one kind or plural kinds of metal atoms and a ligand in which an oxygen atom from a carboxyl group is bonded to the metal atom, wherein the ligand further has a heterocycle with a nitrogen atom as a hetero atom and chelates the metal atom by the oxygen atom and the nitrogen atom.

### [Claim 8]

An organic-inorganic hybrid material having a metal oxide matrix having one kind or plural kinds of metal atoms and a ligand in which an oxygen atom from a carboxyl group is bonded to the metal atom, wherein the ligand further has a carbonyl group, and chelates the metal atom by the oxygen atom and the carbonyl group.

### [Claim 9]

An organic-inorganic hybrid material having a metal oxide matrix having one kind or plural kinds of metal atoms, and a ligand in which an oxygen atom from a carboxyl group is bonded to the metal atom, wherein the ligand further has an azomethine group, and chelates the metal atom by the oxygen atom and the azomethine group.

### [Claim 10]

An organic-inorganic hybrid material having a metal oxide matrix having one kind or plural kinds of metal atoms and a ligand in which an oxygen atom from a hydroxylamino group is bonded to the metal atom, wherein the ligand further has a carbonyl group and chelates the metal atom by the oxygen atom and the carbonyl group.

### [Claim 11]

An organic-inorganic hybrid material according to any one of claims 1 to 3, wherein a ligand having any structure of 8-hydroxyquinoline and derivatives thereof, 10-hydroxybenzo[h]-quinoline and derivatives thereof, 2-(2-hydroxyphenyl)benzoxazole and derivatives thereof, 2-(2-hydroxyphenyl)benzothiazole and derivatives thereof, 2-(2-hydroxyphenyl)pyridine and derivatives thereof, 3-hydroxyflavone and derivatives thereof, 5-hydroxyflavone and derivatives thereof, salicylideneamine and derivatives thereof, picolinic acid and

derivatives thereof, coumarin-3-carboxylic acid and derivatives thereof, salicylideneamine acid and derivatives thereof, benzylideneamino acid and derivatives thereof, N-benzoyl-N-phenyl-hydroxylamine and derivatives thereof, and N-cynnamoyl-N-phenyl-hydroxylamine and derivatives thereof is used as the ligand.

### 5 [Claim 12]

The organic-inorganic hybrid material described in any one of claims 1 to 11, wherein the metal atom is any of selected from the group consisting of magnesium, calcium, strontium, barium, scandium, yttrium, lanthanum, titanium, zirconium, hafnium, zinc, aluminum, gallium, and indium.

### 10 [Claim 13]

The organic-inorganic hybrid material described in any one of claims 1 to 12, wherein an aromatic compound is further added to the organic-inorganic hybrid.

### [Claim 14]

The organic-inorganic hybrid material described in claim 13, wherein the aromatic compound is an organic pigment, an organic light emitter, or an organic semiconductor.

### [Claim 15]

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A carrier-injection type electroluminescence element in which the organic-inorganic hybrid material described in any one of claims 1 to 14 is used.

### [Claim 16]

An intrinsic electroluminescent element in which the organic-inorganic hybrid material described in any one of claims 1 to 14 is used.

### [Claim 17]

A light-emitting device using the electroluminescent device described in claim 15 or 16.

## 25 [Claim 18]

A glass product coated with the organic-inorganic hybrid material described in any one of claims 1 to 14.

### [Claim 19]

A composition for coating application, including at least a metal alkoxide having one kind or plural kinds of metal atoms, an organic compound showing a coloring property by chelating the metal atom, and an organic solvent.

### [Claim 20]

A composition for coating application, including at least a metal alkoxide having one kind or plural kinds of metal atoms, an organic compound showing a light-emitting property by chelating the metal atom, and an organic solvent.

#### 5 [Claim 21]

A composition for coating application, including at least a metal alkoxide having one kind or plural kinds of metal atoms, an organic compound showing semiconductivity by chelating the metal atom, and an organic solvent.

### [Claim 22]

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A composition for coating application, including at least a metal alkoxide having one kind or plural kinds of metal atoms, an organic compound having a phenolic hydroxyl group and a heterocycle with a nitrogen atom as a hetero atom, and an organic solvent.

#### [Claim 23]

A composition for coating application, including at least a metal alkoxide having one kind or plural kinds of metal atoms, an organic compound having a phenolic hydroxyl group and a carbonyl group, and an organic solvent.

## [Claim 24]

A composition for coating application, including at least a metal alkoxide having one kind or plural kinds of metal atoms, an organic compound having a phenolic hydroxyl group and an azomethine group, and an organic solvent.

### [Claim 25]

A composition for coating application, including at least a metal alkoxide having one kind or plural kinds of metal atoms, an organic compound having a carboxyl group and a heterocycle with a nitrogen atom as a hetero atom, and an organic solvent.

#### [Claim 26]

A composition for coating application, including at least a metal alkoxide having one kind or plural kinds of metal atoms, an organic compound having a carboxyl group and a carbonyl group, and an organic solvent.

### [Claim 27]

A composition for coating application, including at least a metal alkoxide having one kind or plural kinds of metal atoms, an organic compound having a carboxyl group and an azomethine group, and an organic solvent.

#### [Claim 28]

A composition for coating application, including at least a metal alkoxide having one kind or plural kinds of metal atoms, an organic compound having a hydroxylamino group and a carbonyl group, and an organic solvent.

### [Claim 29]

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A composition for coating application described in any one of claims 19 to 21, wherein the organic compound is any of 8-hydroxyquinoline and derivatives thereof, 10-hydroxybenzo[h]-quinoline and derivatives thereof, 2-(2-hydroxyphenyl)benzoxazole and derivatives thereof, 2-(2-hydroxyphenyl)benzothiazole and derivatives thereof, 2-(2-hydroxyphenyl)benzimidazole and derivatives thereof, 2-(2-hydroxyphenyl)pyridine and derivatives thereof, 3-hydroxyflavone and derivatives thereof, 5-hydroxyflavone and derivatives thereof, salicylideneamine and derivatives thereof, picolinic acid and derivatives thereof, coumarin-3-carboxylic acid and derivatives thereof, salicylideneamino acid and derivatives thereof, benzylideneamino acid and derivatives thereof, N-benzoyl-N-phenyl-hydroxylamine and derivatives thereof. and N-cynnamoyl-N-phenyl-hydroxylamine and derivatives thereof.

### [Claim 30]

The composition for coating application described in any one of claims 19 to 29, wherein the metal atom is any selected from the group consisting of magnesium, calcium, strontium, barium, titanium, zirconium, hafnium, zinc, aluminum, gallium, and indium.

### [Claim 31]

The composition for coating application described in any one of claims 19 to 30, wherein the amount of the organic compound is 1 equivalent or less to the metal alkoxide.

### [Claim 32]

The composition for coating application described in any one of claims 19 to 31, wherein the organic solvent is an organic solvent including a lower alcohol, tetrahydrofuran, or acetonitrile.

### [Claim 33]

The composition for coating application described in claim 32, wherein the lower alcohol is selected from the group consisting of methanol, ethanol, n-propanol, isopropanol,

n-butanol, sec-butanol, and tert-butanol.

### [Claim 34]

The composition for coating application described in any one of claims 19 to 33, wherein water is further added to the composition.

#### 5 [Claim 35]

The composition for coating application described in claim 34, wherein the amount of the water is 2 equivalents or more and 6 equivalents or less to the metal alkoxide.

#### [Claim 36]

The composition for coating application described in any one of claims 19 to 35, wherein a chemical modifier is further added to the composition.

### [Claim 37]

The composition for coating application described in claim 36, wherein the chemical modifier is a  $\beta$ -diketone.

### 15 [Claim 38]

The composition for coating application described in claims 36 or 37, wherein the amount of the chemical modifier is 0.5 equivalent or more and 6 equivalents or less to the metal alkoxide.

### [Claim 39]

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The composition for coating application described in any one of claims 19 to 38, wherein an aromatic compound is further added to the composition.

### [Claim 40]

The composition for coating application described in claim 39, wherein the aromatic compound is an organic pigment, an organic light emitter, or an organic semiconductor.

#### [Claim 41]

A method of manufacturing an organic-inorganic hybrid material, comprising:

- a first step of performing wet coating of the composition for coating application described in any one of claims 19 to 40; and
- a second step of calcining at a temperature of 100 °C or more and 300 °C or less under an atmospheric pressure or under a reduced pressure.

### [Claim 42]

A method of manufacturing an organic-inorganic hybrid material, comprising:

a first step of performing wet coating of a composition for coating application described in claim 37;

a second step of irradiating ultraviolet light, wherein a wavelength of the ultraviolet light overlaps an ultraviolet absorption spectrum of a state in which the  $\beta$ -diketone chelates the metal atom; and

a third step of calcining at a temperature of 100 °C or more and 300 °C or less under an atmospheric pressure or under a reduced pressure.

#### 10 [Claim 43]

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The method of manufacturing the organic-inorganic hybrid material described in claims 41 or 42, wherein the wet coating is any of a dip coating method, a spin coating, and an inkjet method.

[Detailed Description of the Invention]

[0001]

### [Technical Field]

The present invention relates to an organic-inorganic hybrid material that has a metal oxide matrix and a composition for coating application as a starting material of the organic-inorganic hybrid material, and in addition, relates to a manufacturing method of the organic-inorganic hybrid material.

[0002]

### [Background Art]

As a functional material which relates to photonics, electronics, chemistry, or an organism, an organic-inorganic hybrid material has been actively developed recently.

[0003]

There are some proposed synthetic methods of an organic-inorganic hybrid material. For example, intercalation of an organic polar molecule into a layer-structured inorganic compound is known. By using this method, there are proposed applications such as an application to a photonic memory material or an excimer-fluorescent coating material (refer to Patent Document 1) and an application to an organic EL device (non-Patent Document 1).

[0004]

[Patent Document 1]

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Japanese Patent Laid-Open No. 11-263615

[Non-Patent Document 1]

M. Era, et al., Applied Physics Letters, vol. 65, No. 8, 676-678 (1994)

[0005]

Further, sol-gel method has drawn particular attention as a synthetic method of an organic-inorganic hybrid material. The sol-gel method is one kind of chemical reactions, in which a solid is prepared by a liquid phase reaction, and is a synthetic method, in which a state (sol) where colloid is stable with sufficient fluidity is prepared and transformed to a solid state (gel) that does not have the fluidity by evaporating a solvent and the like. As for details of the sol-gel method like this, knowledge can be obtained from a lot of publications (for example, refer to Non-Patent Document 2). In addition, the sol-gel method has been already put into practical use nowadays as a manufacturing process for various coating films and bulk bodies while attracting attention originally as a technique for manufacturing glass (refer to Non-Patent Document 3).

[0006]

[Non-Patent Document 2]

Sumio Sakka, "Science of Sol-Gel Method", Agune Shouhuu, Tokyo, 4-8 (1988)

[Non-Patent Document 3]

Sumio Sakka, Ceramics, vol. 37, No. 3, 136-142 (2002)

[0007]

Then, when a state of dried gel that is obtained by this sol-gel method is used as a final product, an organic-inorganic hybrid material into which a heat-sensitive organic group or organic compound (that is, an organic moiety) is introduced can be easily synthesized with heat treatment at low temperatures from 100 to 200 °C. In other words, being able to form an inorganic matrix (in particular, an oxide matrix) at low temperatures can be said to be one of advantages of the sol-gel method in synthesizing an organic-inorganic hybrid material.

[0008]

Now, in the case of synthesizing an organic-inorganic hybrid material by the

sol-gel method, miscibility between an inorganic matrix and an organic moiety is important. In other words, in order to avoid the aggregation and phase separation of the organic skeleton, it is required that some sort of interaction is given between the inorganic matrix and the organic moiety to disperse uniformly.

[0009]

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As a method for that purpose, it is generally accepted that organosilicate is used to prepare organic-inorganic hybrid. The organosilicate is a silicate in which alkoxy groups are partially substituted by an organic group, which is typified by polydimethylsiloxane. Since the hydrolysis and polycondensation of such an organosilicate allow the formation of an organic-inorganic hybrid material in which an inorganic matrix and an organic moiety are bonded (have interaction) through a covalent bond, a phenomenon such as the foregoing phase separation does not occur.

[0010]

An organic-inorganic hybrid material that can be obtained from such an organosilicate has characteristics of an inorganic compound such as excellent heat resistance and characteristics of an organic compound such as flexibility simultaneously. For example, an organic-inorganic hybrid material that has properties like rubber and excellent heat resistance is synthesized by using polydimethylsiloxane (refer to Non-Patent Document 4). Further, there are applications such as an application to a sliding member (refer to Patent Document 2).

[0011]

[Non-Patent Document 4]

Noriko Yamada, et al., Journal of Materials Chemistry, vol. 7, No. 8, 1491-1495 (1997)

[Patent Document 2]

Japanese Patent Laid-Open No. 2002-212422

[0012]

In addition, there is an organic-inorganic hybrid material synthesized by sol-gel method while an organic compound is further added to the organosilicate. In this case also, the addition of such an organic compound is effective to avoid the phase separation because an interaction such as  $\pi$ - $\pi$  stacking between an organic group of the organosilicate

and the added organic compound can be given. As applications, an application to a colored glass bottle by adding a pigment (refer to Non-Patent Document 5) and an application to an organic EL device by adding a light-emitter (refer to Non-Patent Documents 6 and 7 and Patent Documents 3 and 4) are proposed. In these applications, it can be recognized that a high mechanical strength and durability of an inorganic compound and various optical properties (absorption and luminescence properties) of an organic compound are combined.

[0013]

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[Non-Patent Document 5]

Organic-Inorganic Hybrid Material (Technical Data Collections) (Technical Information Institute Co., Ltd.), 208-215

[Non-Patent Document 6]

Tony Dantas de Morais, et al., *Advanced Materials*, vol. 11, No. 2, 107-112 (1999) [Non-Patent Document 7]

Monika Schneider, et al., *Advanced Materials*, vol. 12, No. 5, 351-354 (2000) [Patent Document 3]

Japanese Patent Laid-Open No. 9-279135

[Patent Document 4]

Japanese Patent Laid-Open No. 2000-306669

20 [0014]

As described above, by using the organosilicate, an organic-inorganic hybrid material that has a silica matrix and has characteristics of organic compound and inorganic compound in combination can be synthesized. However, it is a matter of course that these methods can be only applied to an organic-inorganic hybrid material that has a silica matrix.

[0015]

A lot of metal oxides exist, which are superior in aspects such as durability, heat resistance, and mechanical characteristics. Further, sol-gel method using a silicate is not useful from the viewpoint of a process because it needs an acid or an alkali basically and takes a long time to complete the gelation. Consequently, it is important to discover a method of synthesizing an organic-inorganic hybrid material that has a metal oxide matrix

other than silica matrix.

[0016]

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A method itself of manufacturing a metal oxide by so-gel method, in addition to an organic-inorganic hybrid material, is well known, where a metal alkoxide is frequently used as a starting material. This method has an advantage that an acid or alkali is unnecessary unlike the case of using a silicate. However, since a metal alkoxide has a fairly high rate of hydrolysis (refer to Non-Patent Document 8), it is a drawback to generate precipitation such as a hydroxide or an oxide as soon as water for hydrolysis and polycondensation is added, which inhibits the preparation of stable sol.

[0017]

[Non-Patent Document 8]

Jianye Wen, et al., *Chemistry of Materials*, No. 8, 1667-1681 (1996) [0018]

In order to overcome this drawback, it is necessary to decelerate the hydrolysis rate. As a well-known method, a chemical modification (for example, chelate stabilization) is kwon (refer to Non-Patent Document 9), which prevents a rapid formation of a network due to hydrolysis by partially substituting a part of an alkoxy group of a metal alkoxide with a  $\beta$ -diketone, an alkanol amine, or the like. According to the chemical modification like this, a coating film of a metal oxide material such as zirconia that has a superior strength can be formed by sol-gel method (refer to Patent Document 5).

[0019]

[Non-Patent Document 9]

C. Sanches, et al., *Journal of Non-Crystalline Solids*, vol. 100, 65-76 (1998)
[Patent Document 5]

Japanese Patent Laid-Open No. 10-259095

In addition, there is also an example where a property of a coordinating  $\beta$ -diketone is applied to patterning of a metal oxide (refer to Non-Patent Document 10 and Patent Document 6). Namely, since a chelating  $\beta$ -diketone absorbs light in an ultraviolet region and is easily eliminated by irradiating with ultraviolet light corresponding to the absorption, by irradiating sol with ultraviolet light after coating sol stabilized with a

 $\beta$ -diketone, hydrolysis and polycondensation easily take place only in the irradiated portion to gel. Thus, it is possible to accomplish the patterning since the region that is not irradiated does not gel and can be removed by washing with a solvent.

[0021]

5 [Non-Patent Document 10]

Katsuhide Shinmou, et al., *Japanese Journal of Applied Physics*, vol. 33, No. 8B, L1181-L1184 (1994)

[Patent Document 6]

Japanese Patent No. 3343377

10 [0022]

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However, all of these reports relate the methods for preparing a metal oxide by so-gel method and do not relate the synthesis of an organic-inorganic hybrid material that has a metal oxide matrix.

[0023]

[Problems to be Solved by the Invention]

As described above, most of organic-inorganic hybrid materials have a silica matrix while there are few examples that have a metal oxide matrix. It is possible to simply prepare sol in which a metal alkoxide and some sort of organic compound are dissolved in the same solvent to synthesize an organic-inorganic hybrid material by sol-gel method. However, the above-mentioned problem with respect to miscibility comes out since a metal oxide matrix in this case eventually has no organic group. Therefore, an organic-inorganic hybrid material in which an organic group is directly connected to a metal oxide matrix is required.

[0024]

Further, even in the chemical modification (chelate stabilization) using a  $\beta$ -diketone, which is reported in the above-mentioned Non-Patent Document 10 and Patent Document 6, the  $\beta$ -diketone is added to stabilize sol and is eventually eliminated from the matrix by calcination or irradiation of ultraviolet light. In other words, it is impossible to realize an organic-inorganic hybrid material that develops a different function from a mere metal oxide by keeping the  $\beta$ -diketone directly bonded to a metal oxide matrix.

[0025]

Consequently, it is an object of the present invention to provide an organic-inorganic hybrid material that has an organic group directly connected to a metal oxide. In particular, it is an object thereof to provide a functional organic-inorganic hybrid material in which an organic group directly bonded to a metal oxide develops a coloring property, a light-emitting property, or semiconductivity.

[0026]

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In addition, it is an object of the present invention to provide a starting material (a composition for coating application) for synthesizing an organic-inorganic hybrid material according to the present invention. Further, it is an object thereof to provide a method of manufacturing an organic-inorganic hybrid material according to the present invention by using the composition for coating application.

[0027]

[Means for solving the Problem]

A lot of earnest studies of the inventors have led to a finding that an organic-inorganic hybrid material to achieve the objects can be synthesized by adding an organic compound (typically, a fluorescent chelating agent) that shows a coloring property, a light-emitting property, or semiconductivity upon chelating a metal atom to a solution of a metal alkoxide or a chemically modified metal alkoxide to prepare sol, and by applying sol-gel method to the sol.

[0028]

An organic-inorganic hybrid material according to the present invention has a structure based on a distinctly novel idea that an organic group (chelating agent) that is able to develop a coloring property, a light-emitting property, or semiconductivity is bonded to a metal oxide matrix.

[0029]

A structure of the present invention is an organic-inorganic hybrid material that has a metal oxide matrix that has one kind or plural kinds of metal atoms and a ligand bonded to the metal atom by chelating, where the ligand shows a coloring property, a light-emitting property, or semiconductivity by chelating the metal atom. Hereinafter, a ligand that has the above-mentioned function is referred to as a functional chelating agent.

[0030]

Further, as the functional chelating agent, any of an organic compound in which a chelate is formed by a phenolic hydroxyl group and a heterocycle with a nitrogen atom as a hetero atom, an organic compound in which a chelate is formed by a phenolic hydroxyl group and a carbonyl group, an organic compound in which a chelate is formed by a phenolic hydroxyl group and an azomethine group, an organic compound in which a chelate is formed by a carboxyl group and a heterocycle with a nitrogen atom as a hetero atom, an organic compound in which a chelate is formed by a carboxyl group and a carbonyl group, an organic compound in which a chelate is formed by a carboxyl group and an azomethine group, and an organic compound in which a chelate is formed by a hydroxylamino group and a carbonyl group is preferable.

[0031]

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In these functional chelating agents, each of the phenolic hydroxyl group in case that the chelating agent has the phenolic hydroxyl group, the carboxyl group in case that the chelating agent has the carboxyl group, and the hydroxyl group in case that the chelating agent has the hydroxylamino group is subjected to deprotonation and the oxygen atom is bonded to the metal atom. Then, the chelate is formed due to the formation of a coordinate bond by the nitrogen atom of the heterocycle, the azomethine group, or the carbonyl group. These functional chelating agents are useful since a coloring property, a light-emitting property, or semiconductivity can be easily developed by bonding to the metal atom and, what is more, the bonding strength for the metal is strong.

[0032]

Further, 8-hydroxyquinoline any of and derivatives thereof, 10-hydroxybenzo[h]-quinoline and derivatives thereof, 2-(2-hydroxyphenyl)benzoxazole and derivatives thereof, 2-(2-hydroxyphenyl)benzothiazole and derivatives thereof, 2-(2-hydroxyphenyl)benzimidazole and derivatives thereof, 2-(2-hydroxyphenyl)pyridine and derivatives thereof, 3-hydroxyflavone and derivatives thereof, 5-hydroxyflavone and derivatives thereof, salicylideneamine and derivatives thereof, picolinic acid and derivatives thereof, coumarin-3-carboxylic acid and derivatives thereof, salicylideneamino derivatives thereof, benzylideneamino acid and derivatives thereof, *N*-benzoyl-*N*-phenyl-hydroxylamine derivatives and thereof, and N-cynnamoyl-N-phenyl-hydroxylamine and derivatives thereof, is more preferable as the

functional chelating agent.

[0033]

It is preferable that the metal atom of the metal oxide matrix in the organic-inorganic hybrid material according to the present invention is any element selected from the group consisting of magnesium, calcium, strontium, barium, scandium, yttrium, lanthanum, titanium, zirconium, hafnium, zinc, aluminum, gallium, and indium. By using these elements, particularly strong light emitting can be achieved.

[0034]

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Additionally in the present invention, an aromatic compound may be further added to the above-mentioned organic-inorganic hybrid material according to the present invention. As the aromatic compound to be added at this time, it is preferable to be an organic pigment, an organic light emitter, or an organic semiconductor from the standpoint of functionality in the present invention.

[0035]

Here, it is one of features that the organic-inorganic hybrid material according to the present invention can have a light-emitting property and semiconductivity in combination. Accordingly, the present invention also includes a carrier-injection type electroluminescent device using the organic-inorganic hybrid material according to the present invention and a light-emitting device using the electroluminescent device. Obviously, the organic-inorganic hybrid material according to the present invention can be made an insulator depending on an amount of the functional chelating agent. Therefore, an intrinsic electroluminescent device can also be manufactured. In addition, a light-emitting device using the intrinsic electroluminescent device can also be manufactured.

[0036]

Here, the light-emitting device indicates an image display device or light-emitting device using an electroluminescent element as a light-emitting element. In addition, a module that has a connecter, such as an anisotropic conductive film (FPC: Flexible Printed Circuit) [Sic], a TAB (Tape Automated Bonding) tape, or a TCP (Tape Carrier Package), attached to an electroluminescent device, a module that has a printed wiring board provided at the tip of a TAB tape or a TCP, and a module that has an IC (integrated circuit)

directly mounted on an electroluminescent device by a COG (Chip On Glass) method are all included in the light-emitting device.

[0037]

Further, the organic-inorganic hybrid material according to the present invention is useful since coating on glass is easily achieved so that a glass product that is functionally coated (such as coloring or light emitting) can be prepared. Accordingly, the present invention includes a glass product in which a film of the organic-inorganic hybrid material is formed.

[0038]

Here, it is preferable to synthesize the above-mentioned organic-inorganic hybrid material according to the present invention by sol-gel method. Therefore, it is also an important invention to provide sol (a composition for coating application) that is used at this time. Accordingly, a composition for coating application according to the present invention has a structure of a composition for coating application including at least a metal alkoxide that has one kind or plural kinds of metal atoms, the above-mentioned functional chelating agent, and an organic solvent. As a metal element of the metal alkoxide, it is preferable to be any element selected from the group consisting of magnesium, calcium, strontium, barium, titanium, zirconium, hafnium, zinc, aluminum, gallium, and indium.

[0039]

In addition, it is preferable that the amount of the functional chelating agent to be added as the composition for coating application is 1 equivalent or less to the metal alkoxide on the ground that the organic-inorganic hybrid material according to the present invention is required to form the metal oxide matrix.

[0040]

As the organic solvent in the composition for coating application, it is preferable to be an organic solvent including a lower alcohol, tetrahydrofuran, or acetonitrile. At this time, an alcohol selected from the group consisting of methanol, ethanol, n-propanol, isopropanol, n-butanol, sec-butanol, and tert-butanol is more preferable as the lower alcohol.

[0041]

Further, since the composition for coating application according to the present

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invention is stabilized to some extent by the functional chelating agent serving as a chemical modifier (chelating stabilizer), water may be further added. It is preferable that the amount of the added water in this case is 2 equivalents or more and 6 equivalents or less to the metal alkoxide.

[0042]

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Further, in order to further stabilize the composition for coating application not to produce precipitation, a chemical modifier may be further added to the above-mentioned composition for coating application according to the present invention. In this case, it is preferable to use a  $\beta$ -diketone that is a chelating stabilizer as the chemical modifier. The amount of the added chemical modifier is preferably 0.5 equivalents or more and 6 equivalents or less for the metal alkoxide.

[0043]

Moreover, in the present invention, an aromatic compound may be further added to the above-mentioned composition for coating application according to the present invention. As the aromatic compound to be added at this time, it is preferable to be an organic pigment, an organic light emitter, or an organic semiconductor from the standpoint of functionality in the present invention.

[0044]

Now, a manufacturing method of coating the above-mentioned composition for coating application on a base material to form a film of the organic-inorganic hybrid material according to the present invention is also related to the present invention. Accordingly, the present invention also includes a manufacturing method of an organic-inorganic hybrid material, where calcining is carried out at a temperature of 100 °C or more and 300 °C or less under an atmospheric pressure or under a reduced pressure after wet coating of the composition for coating application according to the present invention on a base material.

[0045]

Further, in the case where the composition for coating application according to the present invention includes a  $\beta$ -diketone as a chemical modifier, ultraviolet light with a wavelength that has overlap with an ultraviolet spectrum of a state of the  $\beta$ -diketone chelating the metal may be irradiated and after that, calcining may be carried out at a

temperature of 100 °C or more and 300 °C or less under an atmospheric pressure or under a reduced pressure.

[0046]

In addition, in the above-mentioned manufacturing method, it is preferable to be any of a dip coating method, a spin coating method, and an inkjet method as a method for the wet coating.

[0047]

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[Embodiment Mode of the Invention]

First, a fundamental conception of the present invention will be described with reference to Fig. 1. In Fig. 1, a case where an organic-inorganic hybrid material 102 according to the present invention, which uses an alumina matrix as a metal oxide matrix and 8-quinolinol as a functional chelating agent, is deposited on a base material 101 will be described as an example. Also in the case of using another metal oxide matrix, in the case of using a complex metal oxide matrix that has a plurality of metal elements, or in the case of using another functional chelating agent, the fundamental principle is the same.

[0048]

As shown in Fig. 1, a fundamental structure of the present invention has a state of a functional chelating agent 104 chelating a metal atom of a metal oxide matrix 103 in a pendant shape. Consequently, the functional chelating agent 104 is combined with the metal oxide matrix 103 while keeping the functional chelating agent 104 forming the chelate, and a nonconventional novel property (a light emission of an organic compound due to a fluorescent chelating agent can be obtained while having a sturdy moiety of a metal oxide, and the like) can be expected. In the example shown in Fig. 1, the metal oxide matrix shows a strong emission since 8-quinolinol forms a chelate with aluminum (see, Embodiment 2).

[0049]

For comparison, Fig. 2 shows a conceptual diagram in the case where a luminescent metal complex (Alq<sub>3</sub>) that has three of 8-quinolinol bonded to aluminum is used to form a thin film 202 (such as a deposited film) thereof on a base material 201. This thin film is an aggregate (a molecular thin film) of an Alq<sub>3</sub> molecule 203, and the molecule 203 has only interaction due to a weak intermolecular force with each other. On

the other hand, when a thin film is formed of the organic-inorganic hybrid material shown in Fig. 1 according to the present invention, a luminescence property similar to that of Alq<sub>3</sub> is shown, and what is more, the thin film is superior also in heat resistance, mechanical strength, and the like because the film is formed by the metal oxide matrix, that is, by the covalent bond.

[0050]

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In addition, the organic-inorganic hybrid material according to the present invention does not exist in a state where an organic compound is just dispersed in the metal oxide matrix, but the metal oxide matrix and the functional chelating agent have interaction with each other by a strong bond of chelate coordination. Therefore, a problem such as a phase separation is not caused.

[0051]

In addition, since an organic group of the functional chelating agent has a bond with the metal oxide matrix in the organic-inorganic hybrid material as shown in Fig. 1, the phase separation does not occur even when an aromatic compound is further added to form an organic-inorganic hybrid material. The conception will be described with reference to Fig. 3.

[0052]

Fig. 3 shows a state of the functional chelating agent 104 chelating the metal atom of the metal oxide matrix 103 in the pendant shape as in Fig. 1. In the case where an aromatic compound 301 is further added to this state,  $\pi$ - $\pi$  stacking interaction 302 is generated between the aromatic compound 301 and the functional chelating agent 104 since there is a  $\pi$ -electron in the functional chelating agent 104. Consequently, a phenomenon such as a phase separation can be avoided.

[0053]

Next, as a method for synthesizing the organic-inorganic hybrid material as shown in Fig. 1, a method of preparing sol including a metal alkoxide and a functional chelating agent (a composition for coating application according to the present invention) to be used for synthesizing by sol-gel method will be exemplified. Fig. 4 and Fig. 5 show schemes thereof. In Fig. 4 and Fig. 5, a case of using an aluminum alkoxide as a metal alkoxide and 8-quinolinol as a functional chelating agent, will be described as an example. Also in

the case of using another metal alkoxide, in the case of using a plurality of metal alkoxides, in the case of using a complex metal alkoxide that has a plurality of metal elements, or in the case of using another functional chelating agent, the fundamental principle is the same.

[0054]

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Fig. 4 is a case of using the functional chelating agent also as a chemical modifier (chelating stabilizer). In Fig.4, a solution 403 in which a metal alkoxide 401 and a functional chelating agent 402 are dissolved and reacted in an appropriate organic solvent at a ratio of 2:1 is prepared, which is followed by the hydrolysis, polycondensation, and calcination to obtain an organic-inorganic hybrid material 404 according to the present invention. As for an amount of added water, it is preferable to be 2 equivalents or more and 6 equivalents or less to the metal alkoxide since the metal of the metal alkoxide is usually divalent to hexavalent. However, the hydrolysis is not always necessary.

[0055]

Further, in the method of Fig. 4, 8-quinolinol is used as both of a functional chelating agent and a chemical modifier (chelating stabilizer). Therefore, in the case of reducing the amount of 8-quinolinol, the stabilization ability is significantly decreased. Consequently, as shown in Fig. 5, another chemical modifier may further be added.

[0056]

In other words, Fig. 5 shows a case where a solution 504 in which a metal alkoxide 501, a functional chelating agent 502, and a chemical modifier 503 are dissolved and reacted in an appropriate organic solvent at a ratio of 2:1:2, which is followed by hydrolysis polycondensation, and calcinations to obtain an organic-inorganic hybrid material 505 according to the present invention. As for an amount of added water, as in the above-mentioned case, it is preferable to be 2 equivalents or more and 6 equivalents or less to the metal alkoxide. However, the hydrolysis is not always necessary.

[0057]

Note that, although ethyl acetoacetate is used here as the chemical modifier 503, the chemical modifier is not limited to this in the present invention, and may be any one as long as it stabilizes sol not to produce precipitation and is easily eliminated eventually by calcining or the like. Furthermore, generally, as long as the amount of the added chemical modifier is 0.5 equivalents or more to the metal alkoxide, an effect can be exerted.

Moreover, since the metal of the metal alkoxide is generally hexavalent or less, it is preferable that the amount of the added chemical modifier is 6 equivalents or less.

[0058]

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The above-mentioned method can be applied in a similar way also when the organic-inorganic hybrid material as shown in Fig. 3 is synthesized. In other words, what to do is only adding an aromatic compound further to the above-mentioned sol and synthesizing the organic-inorganic hybrid material by sol-gel method in a similar way.

[0059]

Also, as a process for obtaining the organic-inorganic hybrid material according to the present invention from the sol prepared as described above, gelation and sintering are carried out by calcining at a temperature of 100 °C or more and 300 °C or less under an atmospheric pressure or under a reduced pressure after wet coating of the sol on a base material.

[0060]

In addition, as shown in Fig. 5, in the case where a  $\beta$ -diketone (ethyl acetoacetate in Fig. 5) is added as the chemical modifier, after wet coating of the sol on a base material, gelation may be carried out by irradiating ultraviolet light with a wavelength that has overlap with an ultraviolet spectrum of a state of the  $\beta$ -diketone coordinating the metal atom (in Fig.5, a state of ethyl acetoacetate coordinating aluminum) to eliminate the  $\beta$ -diketone. Then, after that, the organic-inorganic hybrid material according to the present invention can be obtained by calcining at a temperature of 100  $^{\circ}$ C or more and 300  $^{\circ}$ C or less under an atmospheric pressure or under a reduced pressure.

[0061]

Here, a dip coating method, a spin coating method, and an inkjet method can be used as the above-mentioned wet coating method, which is, however, not limited to these.

[0062]

Next, materials that can be used for manufacturing the organic-inorganic hybrid material according to the present invention will be specifically exemplified below.

[0063]

First, as the functional chelating agent, any of an organic compound in which a chelate is formed by a phenolic hydroxyl group and a heterocycle with a nitrogen atom as a

hetero atom, an organic compound in which a chelate is formed by a phenolic hydroxyl group and a carbonyl group, an organic compound in which a chelate is formed by a phenolic hydroxyl group and an azomethine group, an organic compound in which a chelate is formed by a carboxyl group and a heterocycle with a nitrogen atom as a hetero atom, an organic compound in which a chelate is formed by a carboxyl group and a carbonyl group, an organic compound in which a chelate is formed by a carboxyl group and an azomethine group, and an organic compound in which a chelate is formed by a hydroxylamino group and a carbonyl group is preferable.

[0064]

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As the organic compound in which a chelate is formed by the phenolic hydroxyl group and the heterocycle with the nitrogen atom as the hetero atom, 8-hydroxyquinoline and derivatives thereof shown by the following structure formulas (1) to (8) are representative. In addition, another compounds include 10-hydroxybenzo[h]—quinoline (the following structure formula (9)), 2-(2-hydroxyphenyl)benzoxazole (the following structure formula (10)), 2-(2-hydroxyphenyl)benzothiazole (the following structure formula (11)), a derivative of 2-(2-hydroxyphenyl)benzimidazole (the following structure formula (12) and (13)), 2-(2-hydroxyphenyl)pyridine and a derivative thereof (the following structure formulas (14) and (15)), and the like. Further, compounds such as quinoxalines, phenazines, and naphthyridines, shown by the following structure formulas (16) to (18), can also be applied. However, the organic compound is not limited to these in the present invention.

[0065]

[Chemical Formula 1]

$$H_3C$$
 $N$ 
 $OH$ 
 $OH$ 

[Chemical Formula 2]

[Chemical Formula 3]

[0066]

As the organic compound which forms a chelate by the phenolic hydroxyl group and the carbonyl group, 3-hydroxyflavone (the following structure formula (19)), 5-hydroxyflavone (the following structure formula (20)), and the like are representative. Further, the compounds such as acetophenones and benzophenones, represented by the following structure formulas (21) and (22), can also be applied. However, the organic compound is not limited to these in the present invention.

### [0067]

### [Chemical Formula 4]

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As the organic compound which forms a chelate by the phenolic hydroxyl group and the azomethine group, derivatives of salicylideneamine shown by the following structure formulas (23) to (27) are representative. Further, a salicylideneamine dimmer can be used as shown by the following structure formulas (28) to (31). However, the organic compound is not limited to these in the present invention.

[0069]

[Chemical Formula 5]

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OH

$$CH_3$$
 $CH_3$ 
 $CH_3$ 

[Chemical Formula 6]

OH HO

$$HC=N$$
 $N=CH$ 
 $H_2C-CH_2$ 

(28)

$$HC=N$$
 $N=CH$ 
 $CH_3$ 
 $(29)$ 

As the organic compound in which a chelate is formed by the carboxyl group and the heterocycle with the nitrogen atom as the hetero atom, picolinic acid and derivatives thereof shown by the following structure formulas (32) to (34) are representative. Further, compounds such as pyridines, represented by the following structure formula (35), can also be applied. However, the organic compound is not limited to these in the present

invention. In the present invention, as shown by the following structure formulas (32) to (35), the structure in which the carboxyl group is directly bonded to a conjugated carbon atom is preferable from the standpoints of a light-emitting property and semiconductivity.

## [0071]

## 5 [Chemical Formula 7]

As the organic compound in which a chelate is formed by the carboxyl group and

the carbonyl group, coumarin-3-carboxylic acid (the following structure formula (36)) is representative. Further, chromone-3-carboxylic acid (the following structure formula (37)) and the like can also be applied. However, the organic compound is not limited to these in the present invention. In the present invention, as shown by the following structure formulas (36) and (37), the structure in which the carboxyl group is directly bonded to a conjugated carbon atom is preferable from the standpoints of a light-emitting property and semiconductivity.

[0073]

### [Chemical Formula 8]

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As the organic compound in which a chelate is formed by the carboxyl group and the azomethine group, salicylideneamino acids represented by the following structure formulas (38) to (40) (Since a metal is coordinated also by a phenolic hydroxyl group, these are also the above-mentioned organic compound in which a chelate is formed by the phenolic hydroxyl group and the azomethine group) and benzylideneamino acids represented by the following structure formulas (41) to (43) are representative. However, the organic compound is not limited to these in the present invention. In the present invention, as shown by the following structure formulas (38) to (43), the structure in which

an aromatic ring is bonded to a carbon atom of the azomethine group is preferable from the standpoints of a light-emitting property and semiconductivity.

## [0075]

# [Chemical Formula 9]

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[0076]

As the organic compound in which a chelate is formed by the hydroxylamino group and the carbonyl group, N-benzoyl-N-phenyl-hydroxylamine and derivatives thereof (the following structure formulas (44)and (45)),and N-cynnamoyl-N-phenyl-hydroxylamine (the following structure formula (46)) derivatives thereof, and the like are representative. However, the organic compound is not limited to these in the present invention. In the present invention, as shown by the following structure formulas (44) to (46), the structure in which the carbonyl group is directly bonded to a conjugated carbon atom or the structure in which the hydroxylamino group is directly bonded to a conjugated carbon atom is preferable from the standpoints of a light-emitting property and semiconductivity.

[0077]

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[Chemical Formula 10]

[0078]

On the other hand, as the metal element of the metal oxide matrix, typical metals and transition metals other than semimetal can be used. In particular, a light-emitting property becomes effective by applying magnesium, calcium, strontium, barium, scandium, yttrium, lanthanum, titanium, zirconium, hafnium, zinc, aluminum, gallium, indium, or the like. On the contrary, when the other transition element is applied, a light-emitting property often tends to be reduced, however, a coloring property becomes effective since a light absorption in the region of visible light occurs due to a d-d transition.

[0079]

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In addition, as a material for forming the metal oxide matrix, on the premise of sol-gel method, metal alkoxides are appropriate as described above, which include kinds such as metal n-propoxide, isopropoxide, n-butoxide, sec-butoxide, and tert-butoxide. Since a liquid phase reaction is used in sol-gel method, it is preferable that these alkoxides are liquid or readily dissolved in an organic solvent.

[0080]

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The organic solvent may be any one as long as the functional chelating agent and the metal alkoxide can be dissolved therein, which includes lower alcohols, tetrahydrofuran (THF), acetonitrile, chloroform, dichloroethane, chlorobenzene, and acetone, and like, and these can be used singularly or by mixture. In particular, considering that water is added for hydrolysis and polycondensation, lower alcohols, THF, and acetonitrile are preferable since these are readily mixed with water. In addition, one of the reasons is that these solvents are readily evaporated by calcinations.

[0081]

As the above-mentioned lower alcohols, methanol, ethanol, n-propanol, isopropanol, n-butanol, sec-butanol, and tert-butanol can be used. As for these alcohols, it is preferable to use one that is similar in kind to the alkoxy group of the metal alkoxide (In other words, n-propanol when n-propoxide is used as the metal alkoxide) from the standpoint of preventing an exchange reaction of the alkoxy group and the standpoint of solubility.

[0082]

In addition, known ones can be used as the chemical modifier, and  $\beta$ -diketones typified by acethylacetone, ethyl acetoacetate, benzoylacetone, and the like are preferable.

[0083]

Furthermore, an aromatic compound can be further added to the organic-inorganic compound material as shown in Fig. 3, and organic pigments, organic light emitters, or organic semiconductors can be used as the aromatic compound in this case. The organic pigments include phthalocyanine derivatives, anthraquinone derivatives, violanthrone, phenolphthalein, malachite green, and the like, which are useful as pigments for coloring. The organic light emitters include phosphorescent materials such as bis(2-phenylpyridinato-N,C<sup>2</sup>)(acetylacetonato) iridium (abbreviation: Ir(ppy)<sub>2</sub>(acac)) and

bis(2-(2'-benzothienyl)pyridinato-N,C<sup>3'</sup>)(acetylacetonato) iridium (abbreviation: Ir(btp)<sub>2</sub>(acac)), in addition to fluorescent materials such as coumarin derivatives, rhodamines, and fluoresceins. The organic semiconductors include hole transporting materials such as 4,4'-bis[N-(3-methylphenyl)-N-phenyl-amino]-biphenyl (abbreviation: TPD), 4,4'-bis[N-(1-naphthyl)-N-phenyl-amino]-biphenyl (abbreviation:  $\alpha$ -NPD), 4,4',4"-tris(N,N-diphenyl-amino)-triphenylamine (abbreviation: TDATA), and 4,4',4"-tris[N-(3-methylphenyl)-N-phenyl-amino]-triphenylamine (abbreviation: MTDATA) and electron transporting materials such as 2-(4-biphenylyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole PBD), (abbreviation: 1,3-bis[5-(p-tert-butylphenyl)-1,3,4-oxadiazole-2-yl]benzene (abbreviation: OXD-7), 3-(4-tert-butylphenyl)-4-phenyl-5-(4-biphenylyl)-1,2,4-triazole (abbreviation: TAZ), 3-(4-tert-butylphenyl)-4-(4-ethylphenyl)-5-(4-biphenylyl)-1,2,4-triazole (abbreviation: p-EtTAZ), bathophenanthroline (abbreviation: BPhen), and bathocuproine (abbreviation: BCP).

[0084]

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Note that the application range of the organic-inorganic hybrid material of the present invention is represented by the application to a colored glass and a fluorescent glass which are obtained by the application onto a glass. Furthermore, another application example is given by the application to an electroluminescence element. An embodiment mode of the electroluminescence element will be described in detail below.

[0085]

## [Embodiment Mode 1]

In the Embodiment Mode 1, a structure of a carrier-injection type electroluminescent device using an organic-inorganic hybrid material according to the present invention will be described with reference to Fig. 6(a). Although a substrate is omitted in Fig. 6(a), the substrate may be on any side of an anode 601 and a cathode 603.

[0086]

Fig. 6(a) shows an electroluminescent device that has a structure in which a light-emitting layer 602 containing an organic-inorganic hybrid material according to the present invention is interposed between the anode 601 and the cathode 603. On this occasion, it is preferable to further add a hole transporting material mentioned above in the

case where a functional chelating agent bonded to a matrix shows an electron transporting property. Also, it is preferable to further add an electron transporting material mentioned above in the case where the functional chelating agent bonded to the matrix shows a hole transporting property. This is because adding those additives makes it possible to improve a carrier-injection balance and a carrier-transporting balance, which contributes to improvement of an emission efficiency. Further, an organic light emitter (a fluorescent material or a phosphorescent material) mentioned above may be added to extract a light emission thereof. Obviously, a light emission from the functional chelating agent may be extracted.

[0087]

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Additionally, since many of metal oxide matrixes are electrically non-conductive while the functional chelating agent shows semiconductivity (i.e. take a role of transporting carriers), the light-emitting layer 602 becomes an insulator when the amount of the functional chelating agent is reduced. Therefore, in the case of applying the organic-inorganic hybrid material according to the present invention to a carrier-injection type electroluminescent device as in the Embodiment Mode 1, it is preferable that the proportion of the functional chelating agent to a metal atom is 0.1 equivalent or more.

[8800]

It is preferable to use a conductive material that has a large work function as a material of the anode 601. When light is extracted in a direction to the anode side, a transparent conductive material such as indium-tin oxide (ITO) or indium-zinc oxide (IZO) may be used. When the anode side is made for light shielding, in addition to a single layer film such as TiN, ZrN, Ti, W, Ni, Pt, or Cr, a lamination structure of a titanium nitride film and a film containing aluminum as its main component, and a three-layer structure of a titanium nitride film, a film containing aluminum as its main component, and a titanium nitride film can be used. Alternatively, a method of laminating the above-mentioned transparent conductive material on a reflective electrode such as Ti or Al is applicable.

[0089]

In addition, it is preferable to use a conductive material that has a small work function as a material of the cathode 603, and specifically, in addition to an alkali metal such as Li or Cs, an alkali earth metal such as Mg, Ca, or Sr, and alloys including these

(such as Mg-Ag or Al-Li), it is also possible to form the cathode 603 by using a rare earth metal such as Yb or Er. In addition, in the case of using an electron injecting layer such as LiF, CsF, CaF<sub>2</sub>, or Li<sub>2</sub>O, an usual conductive thin film such as aluminum can be used. In addition, in the case that light is extracted in a direction to the cathode side, a laminated structure of an ultra thin film including an alkali metal such as Li or Cs and an alkali earth metal such as Mg, Ca, or Sr, and a transparent conductive film (such as ITO, IZO, or ZnO) may be used. Alternatively, either an alkali metal or an alkali earth metal and an electron transporting material may be co-evaporated to form an electron injecting layer, and a transparent conductive film (such as ITO, IZO, or ZnO) may be laminated thereon.

[0090]

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Between the anode 601 and the light-emitting layer 602, a hole injecting and transporting layer may be inserted. As materials that can be used for the hole injecting and transporting layer, the above-mentioned hole transporting materials can be used. In addition, porphyrin compounds are efficient among organic compounds, and phthalocyanine (abbreviation: H<sub>2</sub>-Pc), copper phthalocyanine (hereinafter, referred to as CuPc), and the like can be used. A material of a conductive polymer chemically doped is also available, and polyethylenedioxythiophene (abbreviation: PEDOT) doped with poly(styrenesulfonic acid) (abbreviation: PSS), polyaniline (abbreviation: PAni), polyvinylcarbazole (abbreviation: PVK), and the like can be used. Further, a thin film of an inorganic semiconductor such as vanadium pentoxide and an ultra thin film of an inorganic insulator such as aluminum oxide are also efficient.

[0091]

Further, between the cathode 603 and the light-emitting layer 602, an electron injecting and transporting layer may be inserted. As materials that can be used for the electron injecting and transporting layer, the above-mentioned electron transporting materials can be used. In addition, an ultra thin film of an insulator such as an alkali metal halide such as LiF or CsF, an alkali earth halide such as CaF<sub>2</sub>, or an alkali metal oxide such as Li<sub>2</sub>O is often used. Further, alkali-metal complexes such as lithium acetylacetonate (abbreviation: Li(acac)) or 8-quinolinolato-lithium (abbreviation: Liq) are also efficient.

[0092]

In the present invention, a layer interposed between an anode and a cathode, which may include a light-emitting layer, a hole injecting and transporting layer, and an electron injecting and transporting layer, is referred to as an electroluminescent layer.

[0093]

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The thus obtained electroluminescent device is able to achieve light emissions in various colors by changing the type of the functional chelating agent or the type of the organic light emitter added. In addition, the structure formula (1), the structure formula (8), and the structure formula (14) emit light respectively in yellow-green, red-orange, and blue, by chelating. Accordingly, a white light emission can also be achieved easily by mixing these at an appropriate ratio.

[0094]

## [Embodiment Mode 2]

In the Embodiment Mode 2, a structure of an intrinsic electroluminescent device using an organic-inorganic hybrid material according to the present invention will be described with reference to Fig. 6(b). Although a substrate is omitted in Fig. 6(b), the substrate may be on any side of a first electrode 611 and a second electrode 613.

[0095]

Fig. 6(b) is an electroluminescent device that has a structure in which a light-emitting layer 612 comprising an organic-inorganic hybrid material according to the present invention is interposed between the first electrode 611 and the second electrode 613. An organic light emitter (a fluorescent material or a phosphorescent material) as mentioned above may be added to the light-emitting layer 612 to extract a light emission thereof. Obviously, a light emission from a functional chelating agent may be extracted.

[0096]

A different point from Fig. 6(a) is that the light-emitting layer 612 is made an insulator to avoid current from flowing and a light emission due to collision excitation (that is, intrinsic electroluminescence) is derived by driving the device with the used of a means 614 for applying alternating current bias. On this occasion, in order to make the light-emitting layer 612 the insulator, contrary to Embodiment Mode 1, it is preferable that the population of the functional chelating agent to a metal atom is 0.1 equivalent or less.

[0097]

Further, a layer of a dielectric (such as barium titanate) that has a high dielectric constant may be formed between the first electrode 611 and the light-emitting layer 612 or between the second electrode 613 and the light-emitting layer 612.

[0098]

5 [Embodiment]

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[Embodiment 1] (Preparation of sol)

In the present embodiment, a method of preparing a composition for coating application (sol) according to the present invention will be exemplified specifically, where aluminum-sec-butoxide is used as a metal alkoxide, 8-quinolinol is used as an organic compound that shows a coloring property, a light-emitting property, or semiconductivity by chelating a metal atom, isopropanol is used as an organic solvent, and ethyl acetoacetate is used as a chemical modifier.

[0099]

First, in a glove box with relative humidity kept at 5% or less, 0.493 g (2 mmol) of aluminum-sec-butoxide (manufactured by Tokyo Kasei Kogyo Co., Ltd.) was dispersed in 2.404 g (40 mmol) of dehydrated isopropanol (manufactured by Kanto Chemical Co., Inc). Next, a solution of a chemically modified metal alkoxide was prepared by dropping 0.390 g (3 mmol) of ethyl acetoacetate (manufactured by Kishida Chemical Co., Ltd.) while stirring.

[0100]

Separately from this, in the glove box, a solution of 8-quinolinol was prepared by dissolving 0.145 g (1 mmol) of 8-quinolinol (manufactured by Tokyo Kasei Kogyo Co., Ltd.) in 6.010 g (100 mmol) of dehydrated isopropanol and adding 0.090 ml (about 5 mmol) of pure water.

[0101]

Then, the thus obtained solution of 8-quinolinol was dropped in the above-mentioned solution of the metal alkoxide while stirring. On dropping, the solution changed in color from transparent and colorless to yellow. By additional stirring for 1 hour after the dropping, a composition for coating application (sol) according to the present invention was obtained. Precipitation as observed in a usual synthesis of a metal complex was not observed at all. The manufactured sol composition has a ratio of

aluminum-sec-butoxide: 8-quinolinol: ethyl acetoacetate: water: isopropanol = 2:1:3:5:140 [unit; mmol].

[0102]

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The thus obtained composition for coating application (sol) according to the present invention was subjected to measurement of IR absorption spectrum by a liquid As a measurement apparatus, an infrared spectrophotometer membrane technique. (manufactured by Thermo Nicolet Corp.) was used. The result is shown in Fig. 10 [Sic]. In Fig. 10 [Sic], while many of peaks are attributed to isopropanol that is the solvent, peaks are observed in the vicinity of 1610 cm<sup>-1</sup> of C-O stretching vibration in an enol form and in the vicinity of 1530 cm<sup>-1</sup> of C=C stretching vibration of a six-membered ring due to a formation of a chelate ring, and 1730 cm<sup>-1</sup> of C=O stretching vibration in a keto form is negligibly observed. Consequently, the result suggests that the ethyl acetoacetate mostly forms a chelate ring (functions as a chemical modification). In addition, since the composition was colored in yellow on dropping the solution of 8-quinolinol as described above, 8-quinolinol obviously forms a chelate ring. From these results, it was revealed that both ethyl acetoacetate and 8-quinolinol chelate aluminum in the sol of the present embodiment.

[0103]

[Embodiment 2]

In the present embodiment, a method of synthesizing an organic-inorganic hybrid material according to the present invention, in which an alumina matrix is chelated by 8-quinolinol, will be exemplified specifically.

[0104]

First, the sol obtained in Embodiment 1 was dropped on a quartz substrate through a 0.45 µm filter to perform spin coating under conditions of 800 rpm and 30 seconds. After the spin coating, drying and calcining were conducted under four kinds of conditions shown in the following Table 1 to obtain four kinds of samples in total. The calcining was conducted under a nitrogen atmosphere in a gas exchange furnace.

[0105]

30 [Table 1]

sample No.	conditions of drying and calcining
(1)	drying by leaving at room temperature under air
(2)	calcining at 80 °C for 1 h
(3)	After calcining under the condition of (2), further calcining at $150^{\circ}\mathrm{C}$ for 1 h
(4)	After calcining under the condition of (3), further calcining at 200 °C for 1 h

[0106]

A UV-Vis spectrophotometer (from JASCO corporation) was used to measure UV-Vis absorption spectra of the respective samples (1) to (4). The results are shown in Fig. 11 [Sic]. As shown in Fig. 11 [Sic], when (2) and (3) are compared, a broad spectrum at 260 to 270 nm in (2) is varied to a sharp spectrum at 260 nm in (3). When 8-quinolinol forms a chelate ring with a metal, absorptions are observed at two regions, from 370 to 380 nm and in the vicinity of 260 nm. On the other hand, when ethyl acetoacetate forms a chelate ring with a metal, absorption is observed in the vicinity of 270 nm (to be described in Comparative Example 1). Consequently, it is considered that aluminum is coordinated by both 8-quinolinol and ethyl acetoacetate at 80 °C or less so that absorptions at 260 nm and 270 nm are overlapped to develop the broad absorption. In addition, it can be concluded that, on the calcining at 150 °C, the chelate coordination of 8-quinolinol is remaining while the chelate coordination of ethyl acetoacetate is mostly dissociated so that the sharp peak in the vicinity of 260 nm is remaining (in fact, the absorption of 370 to 380 nm did not disappear, and the sample is yellow in color, which suggest that the chelating 8-quinolinol is remaining).

## [0107]

It is known that, in sol-gel method, a chemical modifier such as ethyl acetoacetate is eliminated by calcining to form a metal oxide matrix (a bond of metal-oxygen-metal). Consequently, from Fig.11 [Sic], it can be concluded that an alumina matrix is formed and

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that aluminum is coordinated by 8-quinolinol that is a functional chelating agent in the samples (3) and (4) (that is, the conditions of calcining from 150 °C to 200 °C). From the descriptions above, it is found that an organic-inorganic hybrid material according to the present invention is synthesized.

[0108]

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In addition, (3) and (4) are colored in yellow due to the broad absorption overlapping a visible light range (having a peak top of 370 to 380 nm), and accordingly, which means that a colored organic-inorganic hybrid material can be synthesized by applying the present invention.

[0109]

Further, Fig. 12 [Sic] shows result of measuring photoluminescence of (3) and (4), with exciting light of 365 nm. As shown in Fig. 12 [Sic], either sample shows a yellow-green light emission that has a peak in the vicinity of 530 nm. Accordingly, it turns out that a light-emitting organic-inorganic hybrid material can be synthesized by applying the present invention.

[0110]

[Comparative Example 1]

In the present comparative example, conventional sol in the state where 8-quinolinol is absence in the sol of Embodiment 1 (that is, sol using aluminum-sec-butoxide as a metal alkoxide, ethyl acetoacetate as a chemical modifier, and isopropanol as an organic solvent) was prepared, applied, and calcined.

[0111]

First, in the same way as in Embodiment 1, sol of aluminum-sec-butoxide: ethyl acetoacetate: water: isopropanol = 2:3:5:140 [unit; mmol] was prepared. Next, spin coating on a quartz substrate was conducted in the same way as in Embodiment 2, and four kinds of samples in total, dried and calcined under the above-mentioned conditions ((1) to (4)) of Table 1, were obtained.

[0112]

Fig. 13 [Sic] shows results of measuring UV-Vis absorption spectra of the respective samples (1) to (4) in the present comparative example. As shown in Fig. 13 [Sic], absorption in the vicinity of 270 nm, generated by the formation of a chelate ring of

ethyl acetoacetate with a metal, is reduced in intensity with the increase in calcining temperature, which suggests, in other words, ethyl acetoacetate is eliminated with the increase in calcining temperature and almost completely eliminated at 150 °C to 200 °C to give just an amorphous alumina thin film. Accordingly, it was impossible to form an organic-inorganic hybrid material.

[0113]

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In addition, while a chelate ring of ethyl acetoacetate is remaining in the conditions of (1) or (2) (that is, calcining temperature at 80 °C or less), a function such as a coloring property or a light-emitting property was not observed at all.

[0114]

[Embodiment 3]

In the present embodiment, an example of applying an organic-inorganic hybrid material according to the present invention to a carrier-injection type electroluminescent device will be exemplified specifically. As a device structure, a structure is used in which a hole injecting and transporting layer are inserted to the structure shown in Fig. 6(a).

[0115]

First, an anode is formed over a glass substrate that has an insulating surface. ITO that is a transparent conductive film is used as a material to be formed with a film thickness of 110 nm by a sputtering method. The anode is formed with a size of 2mm×2mm.

[0116]

Next, a hole injecting and transporting layer that is approximately 30 nm in thickness is obtained by spin coating of an aqueous solution in which PEDOT and PSS are mixed on the anode and baking at 150 °C. Further, a light-emitting layer is formed by spin coating of the sol described in Embodiment 1 on the hole injecting and transporting layer and baking at 80 °C for 1 hour and 150 °C for 2 hours. The thus formed hole injecting and transporting layer and light-emitting layer function as an electroluminescent layer.

[0117]

Lastly, a cathode is formed. In the present embodiment, an aluminum and lithium alloy (Al-Li) is formed by vacuum deposition with resistance heating to be 100 nm.

[0118]

In this way, a carrier-injection type electroluminescent device according to the present invention is formed. Although an example of forming an anode on a substrate is described in the Embodiment 3, the present invention is not limited to this. It is also possible to form a cathode over a substrate. However, in this case (that is, the case of replacing the anode with the cathode), the electroluminescent layer has a reverse buildup sequence from the case shown in the present embodiment.

[0119]

Further, although the anode is a transparent electrode to have a structure where light generated in the electroluminescent layer is emitted from the anode side in the present embodiment, the present invention is not limited to this and a structure where light is emitted from the cathode side by selecting a suitable material for having a transmittance is also applicable.

[0120]

15 [Embodiment 4]

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In the present embodiment, a glass product that has a surface coated with an organic-inorganic hybrid material according to the present invention will be exemplified specifically.

[0121]

First, the sol shown in Embodiment 1 is prepared to coat a surface by dipping a transparent glass bottle (dip coating). Next, a yellow colored glass bottle can be obtained by baking at 80 °C for 1 hour and at 150 °C for 2 hours. In addition, a yellow-green fluorescent glass bottle can be obtained.

[0122]

A conventional colored glass bottle colored by adding an impurity such as iron has a problem that recycling is difficult. However, a glass bottle as obtained in the Embodiment 4 can be recycled easily since it is possible to isolate the organic-inorganic hybrid material from the surface by a method such as heating at a melting temperature of glass or more.

30 [0123]

[Embodiment 5]

In the present embodiment, a light-emitting device that has a carrier-injection type electroluminescent device according to the present invention in a pixel portion will be described with reference to Fig. 7. Fig. 7(A) is a top view showing the light-emitting device and Fig. 7(B) is a sectional view of Fig. 7(A) cut along A-A'. Reference numeral 701 indicated by a dotted line denotes a source side driver circuit, 702 denotes a pixel portion, and 703 denotes a gate side driver circuit. In addition, reference numeral 704 denotes a sealing substrate and 705 denotes a sealing agent. An inside region 706 surrounded by the sealing agent 705 may be space filled with an inert gas or may be filled with a solid such as a resin.

[0124]

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Reference numeral 707 denotes a connecting wiring for transmitting signals which are input to the source side driver circuit 701 and the gate side driver circuit 703, and receives a video signal, a clock signal, a start signal, a reset signal, and the like from FPC (Flexible Printed Circuit) 708 that serves as an external input terminal. Although only the FPC is shown here, a printed wiring board (PWB) may be attached to the FPC. The light-emitting device in the specification includes not only a light-emitting device body but also a state where an FPC or a PWB is attached thereto.

[0125]

Next, the sectional structure will be explained with reference to Fig. 7(B). The driver circuits and the pixel portion are formed over a substrate 710. Here, the source side driver circuit 701 as the driver circuit portion and the pixel portion 702 are shown.

[0126]

In the source side driver circuit 701, a CMOS circuit is formed by combining an n-channel TFT 723 and a p-channel TFT 724. The TFTs forming the driver circuit may also be formed of a known CMOS circuit, PMOS circuit, or NMOS circuit. The present embodiment mode shows a driver integrated type in which a driver circuit is formed over a substrate. However, such a structure is not always required, and the driver circuit may be formed not on the device substrate but at the outside thereof.

[0127]

The pixel portion 702 is formed of a plurality of pixels, each including a switching TFT 711, a current controlling TFT 712, and a first electrode 713 connected to a

drain thereof electrically. In addition, an insulator 714 is formed to cover an edge of the first electrode 713. Here, a positive photosensitive acrylic resin film is used to form the insulator 714.

[0128]

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Besides, in order to obtain a favorable coverage, the insulator 714 is formed so as to possess a curved surface with a curvature in a top portion or bottom potion. For example, in the case of using positive photosensitive acrylic as a material of the insulator 714, it is preferable that only the top portion of the insulator 714 has a curved surface with a curvature radius (0.2  $\mu$ m to 3  $\mu$ m). In addition, any of a photosensitive negative type that becomes insoluble in an etchant by light and a positive type that becomes soluble in an etchant by light can be used as the insulator 714.

[0129]

On the first electrode 713, an electroluminescent layer 715 and a second electrode 716 are respectively formed. Here, as a material that is used for the first electrode 713 functioning as an anode, it is preferable to use a material that has a large work function. For example, in addition to a single layer such as an ITO (indium tin oxide) film, an indium zinc oxide (IZO) film, a titanium nitride film, a chromium film, a tungsten film, a Zn film, and a Pt film, a laminate of a titanium nitride film and a film including aluminum as its main component, a three-layer structure of a titanium nitride film, a film including aluminum as its main component, and a titanium nitride film, and the like can be used. When a laminated structure is employed, the resistance as a wiring is low, favorable ohmic contact can be taken, and it is possible to function as an anode.

[0130]

Further, for the electroluminescent layer 715, an organic-inorganic hybrid material according to the present invention can be used partially or entirely. Specifically, the structure as mentioned in Embodiment 3 may be applied.

[0131]

In addition, as a material that is used for the second electrode (cathode) 716 formed on the electroluminescent layer 715, a material that has a small work function (Al, Ag, Li, Ca, an alloy thereof, MgAg, MgIn, AlLi, CaF<sub>2</sub>, or CaN) may be used. In the case of transmitting light generated in the electroluminescent layer 715 through the second

electrode 716, it is preferable to use a laminate of a thinned metal thin film and a transparent conductive film (such as ITO (an alloy of indium oxide and tin oxide), an alloy of indium oxide and zinc oxide (In<sub>2</sub>O<sub>3</sub>-ZnO), or zinc oxide (ZnO)) film as the second electrode (cathode) 716.

[0132]

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Further, a structure is obtained by bonding the sealing substrate 704 and the substrate 710 with the sealing agent 705, where an electroluminescent device 717 is equipped in the region 706 surrounded by the substrate 710, the sealing substrate 704, and the sealing agent 705. The present embodiment also includes a structure where the region 706 is filled with the sealing agent 705, in addition to a structure where inert gas (such as nitrogen or argon) is injected.

[0133]

It is preferable to use epoxy resin for the sealing agent 705. In addition, it is desirable that these materials are a material that allows the transmission of moisture or oxygen as less as possible. Further, as a material that is used for the sealing substrate 704, a plastic substrate comprising FRP (Fiberglass-Reinforced Plastics), PVF (polyvinylfluoride), Mylar, polyester, acrylic, or the like can be used besides a glass substrate and a quarts substrate.

[0134]

In this way, the light-emitting device that has the carrier-injection type electroluminescent device according to the present invention can be obtained.

[0135]

[Embodiment 6]

In the present embodiment, a top-emission structured light-emitting device, where light is extracted from the sealing substrate 704 side in the light-emitting device shown in Fig. 7, will be specifically exemplified. Fig. 8(A) shows a schematic view (sectional view) thereof. In Fig. 8(A), the codes of Fig. 7 are cited.

[0136]

In Fig. 8(A), the first electrode 713 is made to be a light-shielding anode and the second electrode 716 is made to be a light-transmitting cathode to form a top-emission structure. Therefore, as the first electrode 713, a single-layered film such as a titanium

nitride film, a chromium film, a tungsten film, a Zn film, and a Pt film, a laminate of a titanium nitride film and a film including aluminum as its main component, a three-layer structure of a titanium nitride film, a film including aluminum as its main component, and a titanium nitride film, and the like can be used. Further, as the second electrode 716, a laminated structure of a thinned metal thin film and a transparent conductive film (such as ITO, IZO, or ZnO) may be used. Here, a titanium nitride film is applied as the first electrode, and a laminated structure of a Mg-Ag alloy thin film and ITO is applied as the second electrode.

[0137]

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In addition, in the light-emitting device of the present embodiment, since the electroluminescent device 717 using an organic-inorganic hybrid material according to the present invention, which emits white light (for example, a structure including a white luminescent organic-inorganic hybrid material as mentioned in Embodiment Mode 1), is used for a display in full colors, a color filter including a coloring layer 811 and a light-shielding layer (BM) 812 (for a simplification, a overcoat layer is not illustrated here) is provided.

[0138]

Further, a transparent protective layer 801 is formed for sealing the electroluminescent device. It is preferable to use an insulating film containing silicon nitride or silicon oxynitride, which can be obtained by a sputtering method (a DC system or a RF system) or a PCVD method, as its main component, a thin film containing carbon as its main component (such as a diamond-like carbon (DLC) film or a carbon nitride (CN) film), or a laminate thereof as this transparent protective layer 801. The use of a silicon target in an atmosphere including nitrogen and argon allows the formation of a silicon nitride film that has a large blocking effect against impurities such as moisture and an alkali metal. Alternatively, a silicon nitride target may be used. Further, a deposition system using remote plasma may be used to form the transparent protective layer. It is preferable to decrease a film thickness of the transparent protective layer as much as possible in order to allow emitted light to pass through the transparent protective layer.

[0139]

Here, for further sealing the electroluminescent device 717, the region 706 in Fig.

7 is filled with not only the sealing agent 705 but also a second sealing agent 802 to bond the sealing substrate 704. This sealing operation may be conducted in an atmosphere of an inert gas. It is preferable to use epoxy resin also for the second sealing agent 802 like the sealing agent 705.

[0140]

[Embodiment 7]

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In the present embodiment, a dual-emission structured light-emitting device, where light is extracted from both the substrate 710 side and the sealing substrate 704 side in the light-emitting device shown in Fig. 7, will be specifically exemplified. Fig. 8(B) shows a schematic view (sectional view) thereof. In Fig. 8(B), the codes of Fig. 7 are cited.

[0141]

Fig. 8(B) has a fundamental structure similar to that in Fig. 8(A). However, what is different from Fig. 8(A) is that a transparent conductive film such as an ITO film or an IZO film is used as the first electrode 713. Here, the dual-emission structured light-emitting device can be realized by using an ITO.

[0142]

Although no color filter is provided to substrate 710 side in Fig. 8(B), a color filter may be provided so that the both sides have full range of colors. In this case, the color filter formed for the substrate 710 side may be provided in the same way that is used in a conventional liquid crystal display device and the like.

[0143]

[Embodiment 8]

In the present embodiment, various electric appliances completed with the use of a light-emitting device that has an electroluminescent device according to the present invention will be described.

[0144]

As electric appliances manufactured with the use of a light-emitting device that has an electroluminescent device according to the present invention, a video camera, a digital camera, a goggle-type display (head mount display), a navigation system, a sound reproduction device (such as an in-car audio system or an audio set), a laptop personal

computer, a game machine, a portable information terminal (such as a mobile computer, a mobile computer, a mobile phone, a portable game machine, or an electronic book), an image reproduction device equipped with a recording medium (specifically, a device equipped with a display device which can reproduce a recording medium such as a digital video disc (DVD) and display the image), and the like can be given. Fig. 8 shows specific examples of these electric appliances.

[0145]

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Fig. 9(A) is a display device, which includes a frame body 9101, a support 9102, a display portion 9103, a speaker portion 9104, a video input terminal 9105, and the like. A light-emitting device that has an electroluminescent device according to the present invention is used for the display portion 9103 to manufacture the display device. The display device includes all devices for displaying information such as for a personal computer, for receiving TV broad casting, and for displaying an advertisement.

[0146]

Fig. 9(B) is a laptop personal computer, which includes a main body 9201, a frame body 9202, a display portion 9203, a keyboard 9204, an external connection port 9205, a pointing mouse 9206, and the like. A light-emitting device that has an electroluminescent device according to the present invention is used for the display portion 9203 to manufacture the laptop personal computer.

[0147]

Fig. 9(C) is a mobile computer, which includes a main body 9301, a display portion 9302, a switch 9303, an operation key 9304, an infrared port 9305, and the like. A light-emitting device that has an electroluminescent device according to the present invention is used for the display portion 9302 to manufacture the mobile computer.

[0148]

Fig. 9(D) is a portable image reproduction device equipped with a recording medium (specifically, a DVD reproduction device), which includes a main body 9401, a frame body 9402, a display portion A 9403, a display portion B 9404, a recording medium (such as DVD) reading portion 9405, an operation key 9406, a speaker portion 9407, and the like. The display portion A 9403 is used mainly for displaying image information while the display portion B 9404 is used mainly for displaying character information, and a

light-emitting device that has an electroluminescent device according to the present invention is used for these display portion A 9403 and display portion B 9404 to manufacture the portable image reproduction device equipped with the recording medium. The image reproduction device equipped with the recording medium further includes a home game machine and the like.

[0149]

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Fig. 9(E) is a goggle-type display (head mount display), which includes a main body 9501, a display portion 9502, an arm portion 9503, and the like. A light-emitting device that has an electroluminescent device according to the present invention is used for the display portion 9502 to manufacture the goggle-type display.

[0150]

Fig. 9(F) is a video camera, which includes a main body 9601, a display portion 9602, a frame body 9603, an external connection port 9604, a remote control receiving portion 9605, an image receiving portion 9606, a battery 9607, a sound input portion 9608, an operation key 9609, an eye piece 9610, and the like. A light-emitting device that has an electroluminescent device according to the present invention is used for the display portion 9602 to manufacture the video camera.

[0151]

Here, Fig. 9(G) is a mobile phone, which includes a main body 9701, a frame body 9702, a display portion 9703, a voice input portion 9704, a voice output portion 9705, an operation key 9706, an external connection port 9707, an antenna 9708, and the like. A light-emitting device that has an electroluminescent device according to the present invention is used for the display portion 9703 to manufacture the mobile phone. The power consumption of the mobile phone can be suppressed by displaying white characters on a black background in the display portion 9703.

[0152]

Fig. 10a is a dual-emission type laptop PC, which includes a keyboard portion 1001, a display portion 1002, and the like. This laptop PC has a feature of enabling both a light emission 1003 toward a front surface and a light emission 1004 toward a rear surface as shown in Fig. 10b, which is achieved by applying a dual-emission structured light-emitting device according to the present invention, for example, as shown in Fig.

8(B), to the display portion 1002. The structure like this makes it possible to view an image and the like by utilizing the light emission toward the rear surface even when the display portion 1002 is closed, as shown in Fig. 10c.

[0153]

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As described above, a light-emitting device that has an electroluminescent device according to the present invention is quite widely applied, and is applicable to electric appliances in all fields.

[0154]

[Effect of the Invention]

By implementing the present invention, it is possible to provide an organic-inorganic hybrid material that has an organic group directly connected to a metal oxide matrix. In particular, it is possible to provide a functional organic-inorganic hybrid material in which an organic group directly connected to a metal oxide matrix develops a coloring property, a light-emitting property, or semiconductivity.

[0155]

In addition, by implementing the present invention, it is possible to provide a starting material (a composition for coating application) for synthesizing an organic-inorganic hybrid material according to the present invention. Further, it is possible to provide a method to manufacture an organic-inorganic hybrid material according to the present invention using the composition for coating application.

[Brief Description of Drawings]

[Fig. 1]

A conceptual diagram of a thin film using an organic-inorganic hybrid material according to the present invention.

25 [Fig. 2]

A conceptual diagram of a conventional molecular thin film.

[Fig. 3]

A conceptual diagram of an organic-inorganic hybrid material according to the present invention, to which an aromatic compound is added.

30 [Fig. 4]

A diagram showing a manufacturing method of an organic-inorganic hybrid

material using a composition for coating application according to the present invention.

[Fig. 5]

A diagram showing a manufacturing method of an organic-inorganic hybrid material using a composition for coating application according to the present invention.

5 [Fig. 6]

A diagram showing a structure of an electroluminescent device according to the present invention.

[Fig. 7]

A schematic diagram of a light-emitting device according to the present invention.

10 [Fig. 8]

A schematic diagram of a light-emitting device according to the present invention.

[Fig. 9]

A diagram showing examples of electric appliances using a light-emitting device according to the present invention.

15 [Fig. 10]

A diagram showing examples of electric appliances using a light-emitting device according to the present invention.

[Fig. 11]

A diagram showing an IR absorption spectrum of a composition for coating application, which is prepared in Embodiment 1.

[Fig. 12]

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A diagram showing an UV-Vis absorption spectrum of an organic-inorganic hybrid material synthesized in Embodiment 2.

[Fig. 13]

A diagram showing a PL spectrum of an organic-inorganic hybrid material synthesized in Embodiment 2.

[Fig. 14]

A diagram showing an UV-Vis absorption spectrum of a conventional metal oxide thin film synthesized in Comparative Example 1.

[Document Name]

Abstract

[Abstract]

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[Problems to be Solved]

An organic-inorganic hybrid material which has an organic group directly bonded to a metal oxide matrix is provided. In particular, a functional organic-inorganic hybrid material in which an organic group directly bonded to a metal oxide matrix shows a coloring property, an emission property, or semiconductivity is provided. Further, a starting material (a composition for coating application) for the preparation of the organic-inorganic hybrid material of the present invention and a synthetic method using the starting material are provided.

[Means for Solving the Problems]

An organic-inorganic hybrid material chelating a metal atom of a metal oxide matrix 103 in a pendant shape is synthesized, where the organic compound (the functional chelating agent) 104 is able to develop a function of a coloring property, a light-emitting property, or semiconductivity by chelating a metal atom. As a synthesis method, sol (a composition for coating application) including a metal alkoxide and a functional chelating agent is prepared and the organic-inorganic hybrid material may be synthesized by sol-gel method.

[Document Name]

Drawings

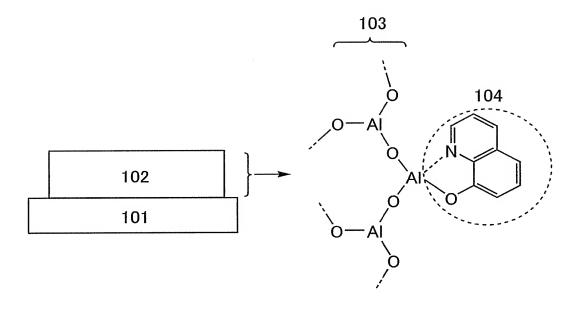


Fig. 1

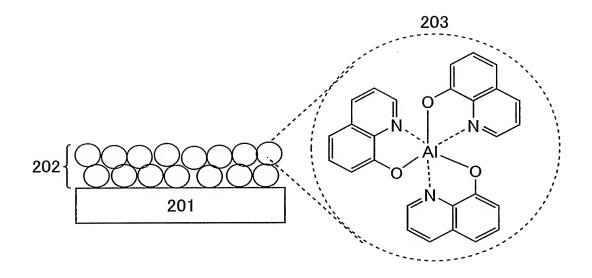


Fig. 2

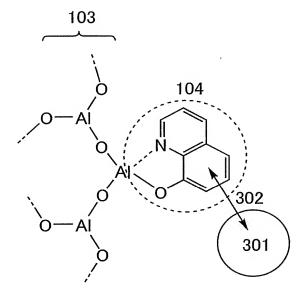
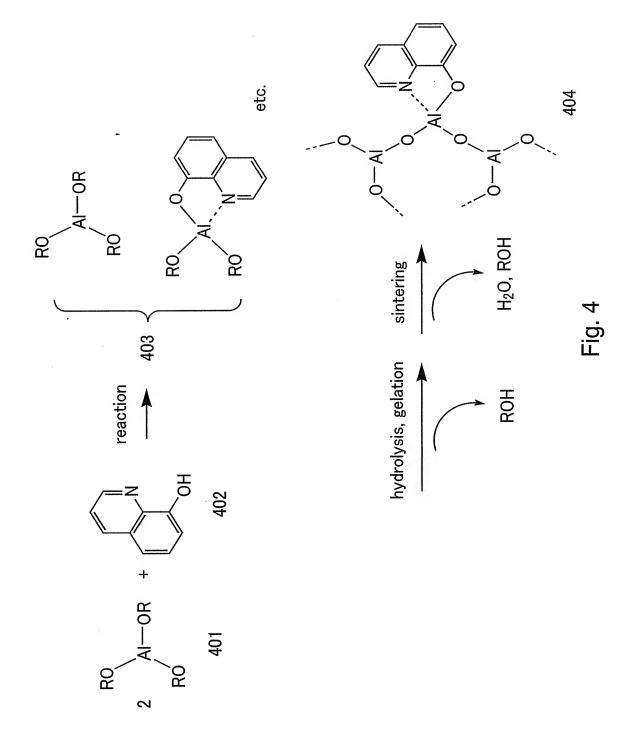
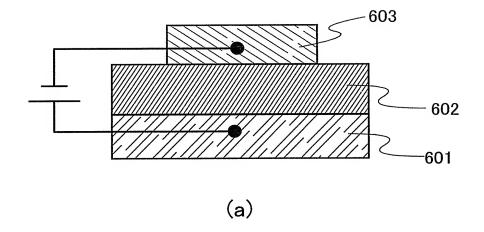


Fig. 3





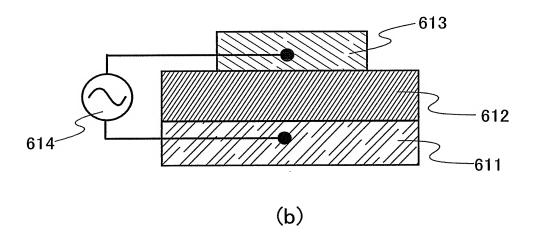


Fig. 6

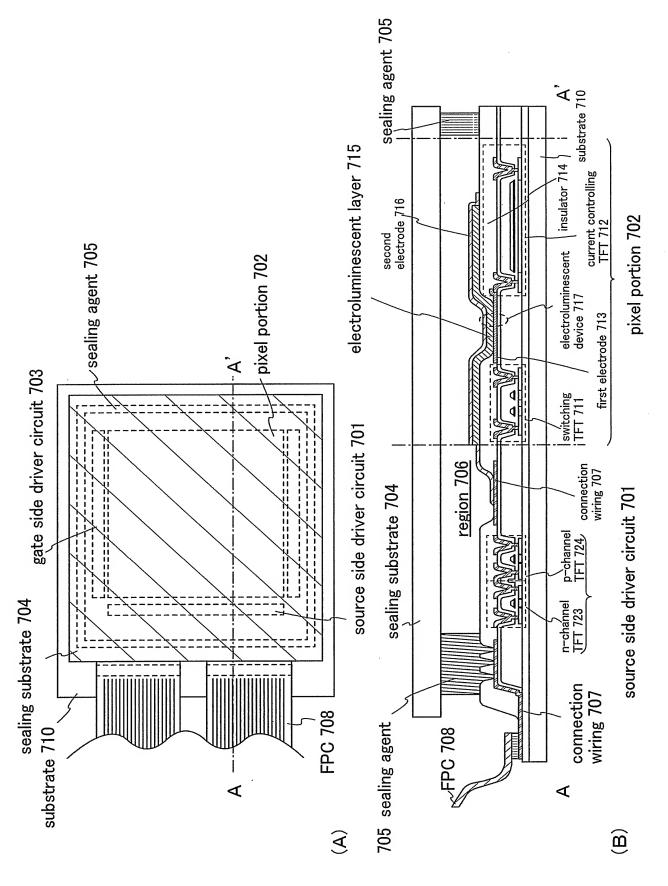


Fig. 7

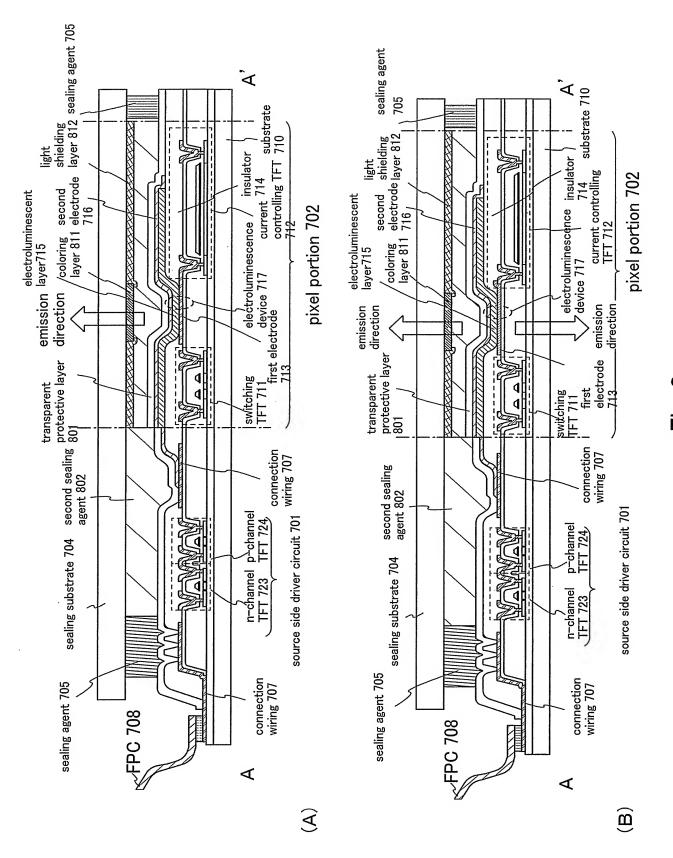
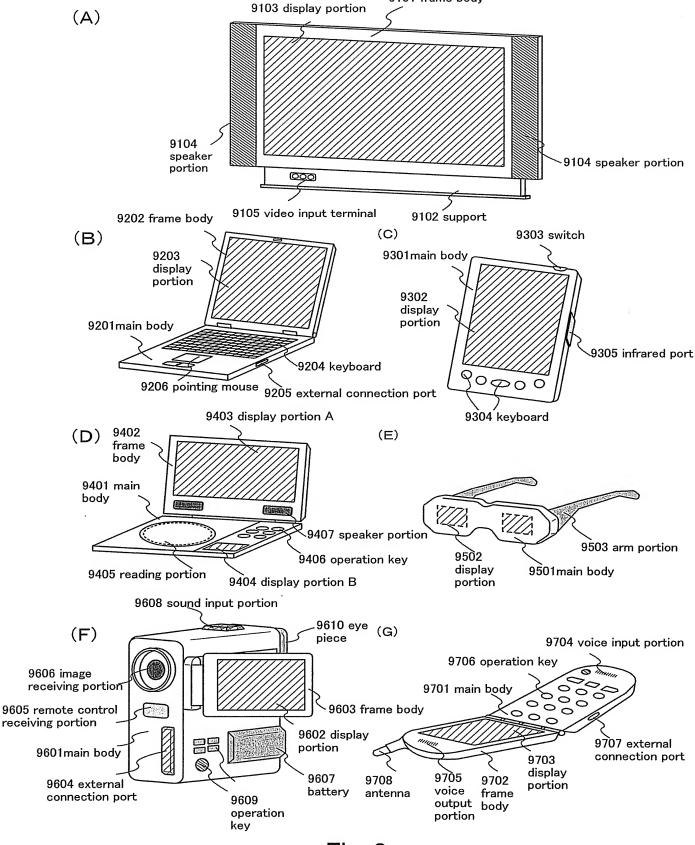
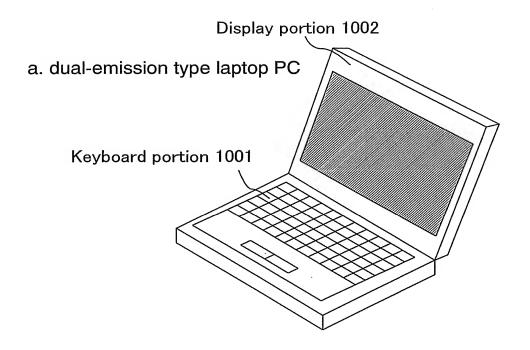


Fig. 8



9101 frame body

Fig. 9



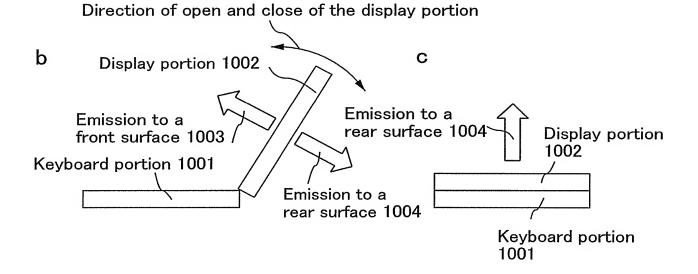


Fig. 10

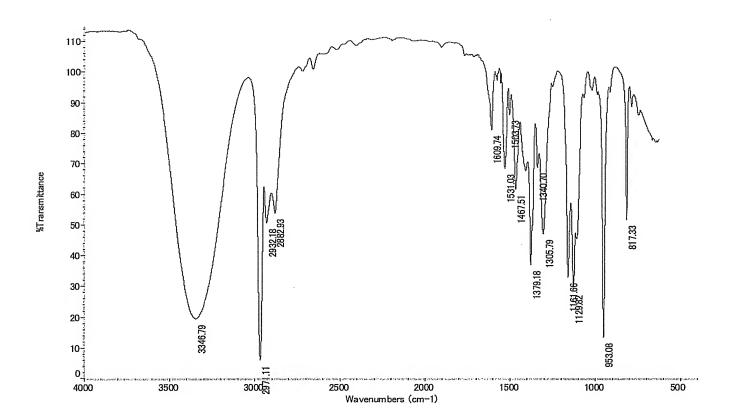


Fig. 11

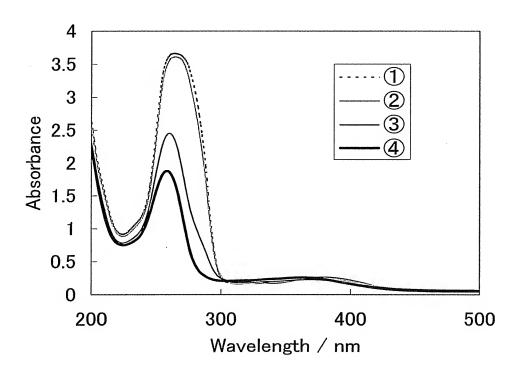


Fig. 12

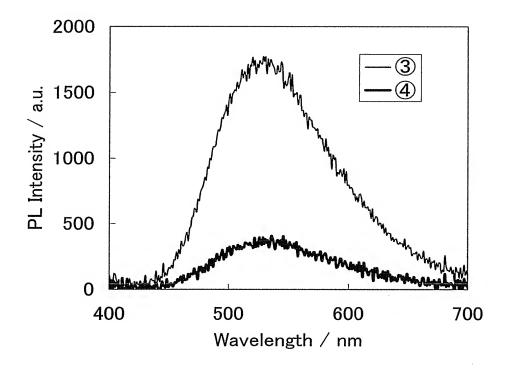


Fig. 13

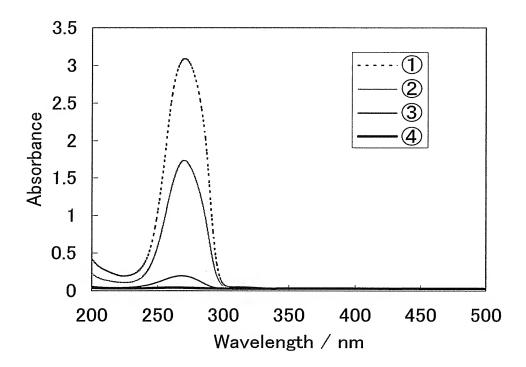


Fig. 14